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Thermodynamic Properties of Coolant Fluids  
and Particle Seeds  
for Gaseous Nuclear Rockets

NASA Contract No. NASw-847

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## FOREWORD

An exploratory experimental and theoretical investigation of gaseous nuclear rocket technology is being conducted by the United Aircraft Corporation Research Laboratories under Contract NASw-847 with the joint AEC-NASA Space Nuclear Propulsion Office. The Technical Supervisor of the Contract for NASA is Captain W. A. Yingling (USAF). Results of the investigation conducted during the period between September 15, 1963 and September 15, 1964 are described in the following nine reports (including the present report) which comprise the required first Interim Summary Technical Report under the Contract:

1. Krascella, N. L.: Theoretical Investigation of the Absorption and Scattering Characteristics of Small Particles. UAC Research Laboratories Report C-910092-1, September 1964. (Unclassified)
2. Marteney, P. J.: Experimental Investigation of the Opacity of Small Particles. UAC Research Laboratories Report C-910092-2, September 1964. (Unclassified)
3. Roback, R.: Thermodynamic Properties of Coolant Fluids and Particle Seeds for Gaseous Nuclear Rockets. UAC Research Laboratories Report C-910092-3, September 1964. (Unclassified) (present report)
4. Schneiderman, S. B.: Theoretical Viscosities and Diffusivities in High-Temperature Mixtures of Hydrogen and Uranium. UAC Research Laboratories Report C-910099-1, September 1964. (Unclassified)
5. McLafferty, G. H.: Analytical Study of Moderator Wall Cooling of Gaseous Nuclear Rocket Engines. UAC Research Laboratories Report C-910093-9, September 1964. (Unclassified)
6. Johnson, B. V.: Analysis of Secondary-Flow-Control Methods for Confined Vortex Flows (U). UAC Research Laboratories Report C-910091-1, September 1964. (Confidential)
7. Travers, A., and B. V. Johnson: Measurements of Flow Characteristics in a Basic Vortex Tube (U). UAC Research Laboratories Report C-910091-2, September 1964. (Confidential)
8. Travers, A., and B. V. Johnson: Measurements of Flow Characteristics in an Axial-Flow Vortex Tube (U). UAC Research Laboratories Report C-910091-3, September 1964. (Confidential)
9. McLafferty, G. H., and W. G. Burwell: Theoretical Investigation of the Temperature Distribution in the Propellant Region of a Vortex-Stabilized Gaseous Nuclear Rocket (U). UAC Research Laboratories Report C-910093-10, September 1964. (Confidential)

Thermodynamic Properties of Coolant Fluids and Particle  
Seeds for Gaseous Nuclear Rockets

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Thermodynamic Properties of Coolant Fluids and Particle

Seeds for Gaseous Nuclear Rockets

SUMMARY

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Thermodynamic properties and equilibrium chemical composition were calculated for various materials which could serve as moderator coolants or as particle seeds designed to control radiant heat transfer in gaseous nuclear rocket engines. The materials which were considered as moderator coolants were hydrogen, methane, water, ammonia, deuterium, heavy water, and helium. Materials which were considered as possible particle seeds were several elemental species which have high boiling points, such as graphite, tungsten, and molybdenum, and the oxides, carbides, nitrides, and borides of titanium and zirconium.

The results of the calculations indicate that the use of graphite as a seed material is impractical because graphite reacts readily with hydrogen, and, consequently, excessive quantities of graphite must be added to the hydrogen stream in order to maintain even very small concentrations of seed particles at high temperatures. Also, the limited thermodynamic data currently available indicate that no significant increase in particle vaporization temperature would result from using oxides, carbides, nitrides, or borides of such high-melting-point metals as titanium and zirconium as seeds in place of the metals themselves.

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INTRODUCTION

As part of the investigation of gaseous nuclear rocket technology conducted under Contract NASw-847, analytical studies were undertaken to investigate the problem of cooling the moderator walls of a gaseous nuclear rocket and to determine the characteristics of seed materials which might be employed in controlling radiant heat transfer to the walls of such a rocket. Since the results of these studies depend on the thermodynamic properties of the materials considered, it was essential that these properties be determined for those conditions of pressure and temperature which are expected to exist in gaseous nuclear rocket engines. Because of the high temperatures which are expected to exist in such an engine, it was necessary that these calculations include the effects of dissociation. The cal-

culations were therefore performed using computational techniques described in Refs. 1 and 2 which have been developed for determining the effect of temperature and pressure on the equilibrium composition and thermodynamic properties of the products of reaction for various chemical systems.

## DISCUSSION

### Properties of Moderator Coolant Fluids

An investigation was conducted to determine the properties of various fluids which might be required in the moderator cooling study described in Ref. 3. The first step in this investigation was to assemble pertinent properties influencing heat transfer for each candidate coolant fluid at a pressure of 1000 atm and a temperature of 4000 R. The data were either obtained from the literature or estimated from various information available in the literature. Data for hydrogen, water, helium, methane, and ammonia at a pressure of 1 atm were obtained directly from Ref. 4. Properties of deuterium and heavy water at a pressure of 1 atm were calculated using the data in Refs. 5 and 6 and a procedure described in Ref. 4 which is based on the Lennard-Jones potential. The viscosity and thermal conductivity at a pressure of 1000 atm were calculated from the data at 1 atm by applying the pressure correction obtained by utilization of the Enskog theory for the viscosity and thermal conductivities of dense gases (Ref. 7). The results of the studies of the properties of the various coolant fluids are presented in Table I. It should be noted that all data shown are for the pure gas with no correction for any deviation from ideality (i.e., for a value of compressibility factor,  $Z$ , equals 1) or for any dissociation which may occur at the temperature and pressure considered.

Since significant dissociation of some of the candidate coolants might occur at the conditions described above, it appeared desirable to determine the extent to which the materials might dissociate over the temperature range likely to be encountered during the operation of nuclear rocket engines. Therefore, a series of calculations were performed using an IBM computation procedure developed at the UAC Research Laboratories and described in Ref. 1 to determine the equilibrium composition of the dissociation products of hydrogen, methane, ammonia, and water at a pressure of 1000 atm and temperatures ranging from 1800 to 10,000 R. All calculations were made using the assumption that the dissociation products behaved as ideal gases.

The calculations were made only for the materials cited above since it can be assumed that the properties of deuterium and heavy water would parallel those of hydrogen and water, and that helium would remain chemically stable at the temperatures considered. Only atomic and molecular products of dissociation were considered

in the calculations because electron and ion concentrations only become significant at temperatures exceeding 10,000 R. The results of the calculations are shown in Figs. 1 through 4.

It can be seen from these figures that dissociation of hydrogen and water becomes significant above 7000 R. On the other hand, appreciable dissociation of methane occurs at a temperature of only 2000 R and ammonia is almost entirely dissociated at 2000 R. Therefore, the properties of methane and ammonia should be substantially different from the estimated values shown in Table I.

It can also be seen from Fig. 2 that a large part of the carbon in methane would exist as a solid at temperatures between approximately 3000 and 6500 R. This solid carbon could very easily clog the coolant passages of a gaseous nuclear rocket. If hydrogen were mixed with methane, this excess hydrogen would tend to shift the equilibrium toward the formation of smaller amounts of solid carbon and, therefore, less clogging of the coolant passages would occur. Additional calculations were made to show the effect of carbon-to-hydrogen weight ratio on the equilibrium composition of solid carbon. The results of these calculations are shown in Fig. 5, and they indicate that no solid carbon, and thus no clogging of the coolant passages, would occur at a temperature of 4500 R for weight ratios of carbon to hydrogen less than approximately 0.73. This ratio corresponds to a mixture of 56.4 per cent methane and 43.6 per cent hydrogen by weight. Such a mixture would also prevent chemical attack on graphite walls by hydrogen at a temperature of 4500 R, but would not prevent this chemical attack at temperatures higher or lower than 4500 R.

The densities of hydrogen, methane, water, and ammonia are shown in Fig. 6 as a function of temperature. Corresponding values of enthalpy (defined as the difference between the total enthalpy of the products of dissociation at a given temperature and the enthalpy of the propellant as a liquid) are shown in Fig. 7.

The ratio of the density and enthalpy of methane, water, and ammonia to those of hydrogen are shown in Fig. 8. These ratios are important since they may serve to determine the effect on performance of a nuclear rocket engine when either water, methane, or ammonia is substituted in whole or in part for hydrogen. For instance, the specific impulse attainable by the use of a given propellant is, in general, proportional to the square root of the exhaust enthalpy of the propellant. For a solid-core nuclear rocket operating at a fixed maximum temperature, the ratio of specific impulse attainable using a given propellant to that attainable using hydrogen is proportional to the square root of the ratio of the enthalpy of this propellant to that of hydrogen at the same operating temperature. Such ratios are noted on the right-hand ordinate of the plot shown in Fig. 8. The maximum enthalpy of the propellant passing through the exhaust nozzle of a gaseous nuclear rocket can be shown (Ref. 3) to be approximately ten times the enthalpy associated with the maximum wall temperature (assuming no space radiators). If this ratio is assumed to be the same for all propellants and if the maximum temperature of the propellant injected into the

cavity of a gaseous nuclear rocket is the same as that for hydrogen, then the specific impulse ratios shown in the right side of Fig. 8 also apply directly for gaseous nuclear rocket engines. According to Fig. 5, a mixture of methane and hydrogen comprising 56.4 per cent methane by weight would prevent any clogging of coolant passages at 4500 R if such a mixture were used as a propellant. From Fig. 8, it can be shown that this mixture would result in a specific impulse at 4500 R of approximately 82 per cent of that of pure hydrogen.

In order to determine the differences in the properties of hydrogen from those of some other material at conditions which are likely to be encountered near the core of a nuclear rocket, additional calculations were made to determine the equilibrium composition and thermodynamic properties of water at temperatures up to 200,000 R and for pressures of 100 and 1000 atm. The calculations were extended for water because it is easily stored and is inexpensive and, for these reasons, might be attractive for initial testing of a gaseous nuclear rocket engine. Also, the properties of methane and ammonia appear to fall between those of hydrogen and water as seen in Figs. 6 and 7, and, therefore, property calculations for both hydrogen and water for very high temperatures and for various pressures would represent extremes of the capability of the four materials which might be utilized as working fluids in a gaseous nuclear rocket engine.

The composition of water at the conditions stated above are shown in Figs. 9 and 10. It can be seen that singly ionized hydrogen and oxygen atoms and the resulting electrons become important at about 30,000 R and are the predominant species at 60,000 R. At higher temperatures, doubly-, triply-, and quadruply-ionized oxygen atoms become important. In calculating the results, no provisions were made for the change in ionization potential of the ionic species due to the presence of electrons.

The density and enthalpy of water at temperatures up to 200,000 R are shown in Fig. 11. The nonlinear variation of enthalpy and density with temperature is due to the dissociation and ionization of the various species as temperature is increased. Also shown in Fig. 11 is the heat content per unit volume (product of enthalpy and density) which is of interest in analyzing the rate of energy removal from a gaseous nuclear rocket for a fixed convective velocity. The composition, density, and enthalpy data presented in Figs. 9, 10, and 11 may be compared with the corresponding data for hydrogen in Ref. 2.

The thermodynamic data (heat capacity, enthalpy, and entropy) for the individual species considered in the calculation were obtained from various sources. Data for the molecular and atomic species for the temperature range up to 6000 K (10,800 R) were obtained from Ref. 8 and were extrapolated to the higher temperatures. Data for atomic hydrogen and atomic oxygen for temperatures exceeding 10,800 R and for all ionic species except the hydroxyl ion and electron were obtained from Ref. 9. Data for electron gas were obtained from Ref. 10 and extrapolated to higher temperatures. Heat capacity and entropy data for the hydroxyl ion were estimated from data for

the hydroxyl radical reported in Ref. 10. The enthalpy data for the hydroxyl ion were calculated using a value of electron affinity obtained from Ref. 11. The hydroxyl ion data were also extrapolated from 6000 K to the higher temperatures. Any errors in thermodynamic data introduced by the extrapolations would probably have little effect on the properties of the mixture as a whole since, in general, the concentrations for those species for which data were extrapolated become insignificant in the temperature range of the extrapolation.

### Chemical Reactions Between Hydrogen and Particle Seeds

Various studies have indicated that solid materials in the form of small particles may be used as propellant seeds in order to control radiant heat transfer in gaseous nuclear rocket engines. The use of solid seeds is usually more desirable than the use of gaseous seeds because the opacity of particles, even if they become liquefied, is continuous over a wide part of the spectrum whereas the opacity of gases exhibits windows at certain wave lengths. Since it is desirable that the particles provide opacity to temperatures approaching 10,000 R, the temperature at which the opacity of hydrogen itself becomes significant (Ref. 2), only those materials with high boiling points were considered as possible seed candidates.

Particles which have been heated by thermal radiation cause heating of the surrounding gas by laminar conduction of the heat to gaseous atoms located in the vicinity of the particle. If the ratio of the thermal conductivity of the gas to the diffusivity of the gas (Lewis number) is on the order of unity, which is the case for most gases, then the diffusion of any vapor formed by the chemical interaction of the particle material and the surrounding gas should occur as rapidly as the conduction of heat from the particle to the surrounding gases. Therefore, it would appear unlikely that non-equilibrium mixtures of reaction products would occur locally and it can be assumed that the particle materials should be in thermodynamic equilibrium with the surrounding gases. A series of calculations of equilibrium composition of the products of reaction of various seed materials with hydrogen was performed and the results of these calculations are discussed in the following subsections.

#### Graphite Particles

According to Ref. 12, graphite particles provide substantially greater opacity per pound than particles made of other materials which have comparable boiling points. However, as can be seen from Fig. 2, hydrogen will react with graphite to form various hydrocarbons along with free hydrogen and carbon. For a given weight ratio of carbon to hydrogen, the largest concentration of free carbon occurs at a temperature of approximately 4500 R (see Fig. 5). Fig. 5 also shows that a weight ratio of carbon to hydrogen greater than 0.73 is required at 4500 R to permit any amount of free carbon and that the amount of free carbon decreases very rapidly as temperature is increased. Since it is desirable that seed materials have the capability of controlling



radiant heat transfer at temperatures up to 10,000 R, it appears that the use of graphite particles as a seed is impractical because of the high carbon-to-hydrogen weight ratios required to provide any amount of solid carbon at this temperature.

It can be seen from Figs. 3 and 4 that the dissociation of water and ammonia leads to the presence of substantial quantities of hydrogen at temperatures of the order of 7000 to 10,000 R. It would be expected that this hydrogen would react with any graphite seed particles in much the same manner as indicated in Figs. 2 and 5, and, therefore, the use of graphite particles for controlling radiant heat transfer in engines employing ammonia or water as working fluids also appears to be impractical.

#### Particles Made From Materials Other Than Graphite

Since the use of graphite as a seed material is impractical because it reacts with hydrogen, a study was undertaken to predict the extent of equilibrium reaction at various temperatures when selected alternate seed materials are added in small amounts to a hydrogen propellant stream. Vapor pressure data from Ref. 13 indicated that many metals have high normal boiling points and would be good candidates for seed materials if their spectral absorption characteristics were good. Calculations were performed using the vapor pressure data from Ref. 13 to determine the amount of condensed metal which will be present in the temperature range 2000 R to 10,000 R if 2 per cent by weight of the material is added to hydrogen at a pressure of 1000 atm. Vapor pressure data was all that was required to determine the state of the metal at each temperature because a literature survey indicated that, although several of the metals do absorb hydrogen to form complex hydrides, these hydrides would decompose at the temperatures considered in this study, and, therefore, it may be assumed that the metals considered will not react with hydrogen propellant in the temperature range of interest. The results of the calculations are shown in Fig. 12.

A series of calculations was also performed to determine the equilibrium composition of the products of reaction of hydrogen with the oxides, nitrides, carbides, and borides of titanium and zirconium. As in the case of the metals, it was assumed that 2 per cent of the seed material was introduced into the hydrogen stream at a pressure of 1000 atm. The results of the calculations indicated that  $\text{TiC}$ ,  $\text{TiN}$ ,  $\text{TiB}_2$ , and  $\text{TiO}_2$  were completely vaporized or dissociated for the conditions described above at temperatures of 6372, 6138, 6210, and 5040 R, respectively. If these vaporization temperatures are compared with that obtained from Fig. 12 for titanium metal itself, namely, 6120 R, it can be seen that the carbide, nitride, and boride offer slight advantages over the parent metal itself. The results of the zirconium calculations indicated that  $\text{ZrC}$ ,  $\text{ZrB}_2$ ,  $\text{ZrN}$ , and  $\text{ZrO}_2$  vaporized at temperatures of 6876, 5760, 6570, and 5472 R, respectively. All these temperatures are considerably lower than the vaporization temperature of zirconium metal (7900 R) obtained from Fig. 12, and, as a result, compounds of zirconium would offer only the advantage of a higher melting temperature than zirconium.

If the trends shown by the vaporization data for the compounds of titanium and zirconium are characteristic of the other high-melting-point metals, it would seem that no significant improvement in the vaporization temperature of particle seeds would be obtained by using the compounds rather than the elemental metal, and that the use of the metals themselves should receive top priority in experimental evaluations of the optical properties of seeded hydrogen streams. At the same time, however, effort should be continued to obtain improved values of the thermodynamic properties (and particularly vaporization data) for carbides, nitrides, borides, and oxides of the high-melting-point metals to provide a firm basis for continuing evaluation of potential seed materials.

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TABLE I

Thermodynamic and Transport Properties of Possible  
Gaseous Nuclear Rocket Moderator Coolant Fluids  
at 4000 R and a Pressure of 1000 Atmospheres

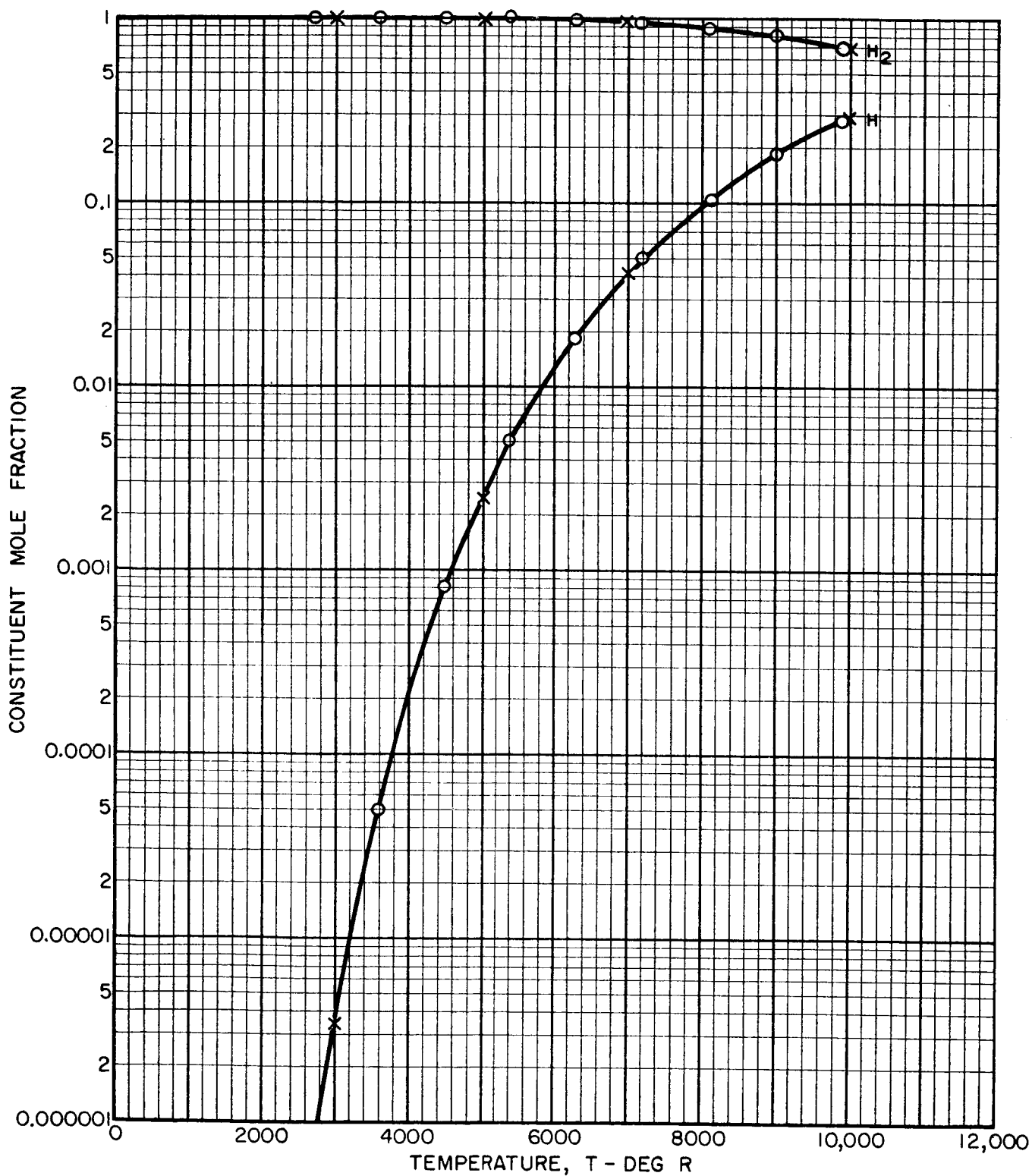
Properties do not include effect of dissociation

Fluid	Density $\frac{(\text{LB})}{(\text{FT}^3)}$	Specific Heat $\frac{(\text{BTU})}{(\text{LB})(\text{DEG R})}$	Viscosity $\frac{(\text{LB})}{(\text{FT})(\text{SEC})}$	Thermal Conductivity $\frac{(\text{BTU})}{(\text{FT}^2)(\text{SEC})(\text{DEG R/FT})}$	Prandtl No.
H <sub>2</sub>	0.6903	4.137	$2.322 \times 10^{-5}$	$14.444 \times 10^{-5}$	0.665
D <sub>2</sub>	1.3793	2.159	$3.197 \times 10^{-5}$	$10.342 \times 10^{-5}$	0.667
H <sub>2</sub> O	6.1686	0.6981	$5.006 \times 10^{-5}$	$5.074 \times 10^{-5}$	0.689
D <sub>2</sub> O	6.8577	0.7120	$5.172 \times 10^{-5}$	$5.269 \times 10^{-5}$	0.699
He	1.3706	1.2411	$5.126 \times 10^{-5}$	$10.023 \times 10^{-5}$	0.635
CH <sub>4</sub> *	5.4930	1.4396	$3.555 \times 10^{-5}$	$7.645 \times 10^{-5}$	0.669
NH <sub>3</sub> *	5.8317	1.0370	$4.495 \times 10^{-5}$	$6.835 \times 10^{-5}$	0.682

\* Dissociation should substantially alter the properties of methane and ammonia and, therefore, the values noted above should be used only as a first estimate (see text and Figs. 2 and 3) or as a base for determining properties at lower temperatures where dissociation is not important.

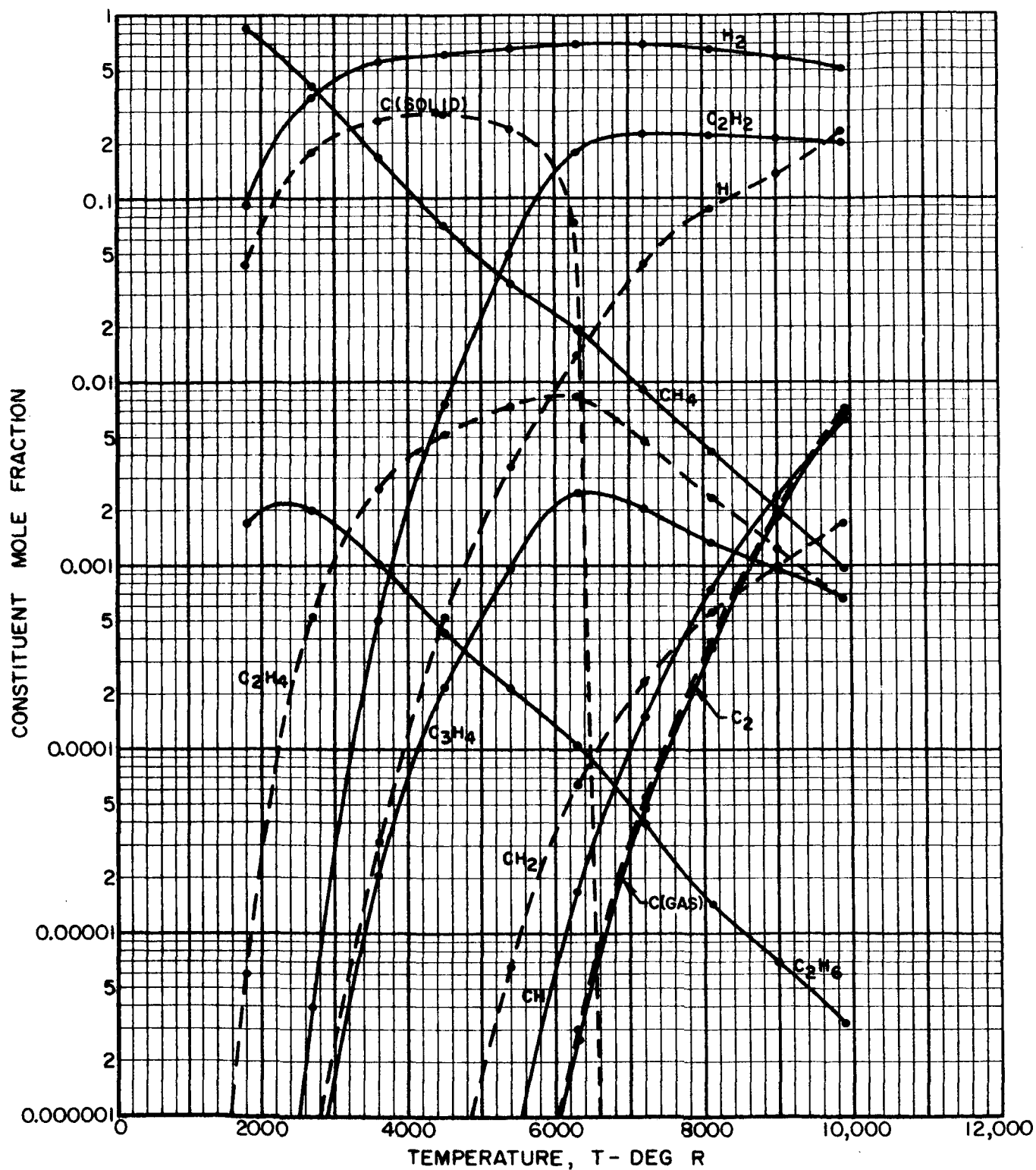
# EFFECT OF TEMPERATURE ON THE COMPOSITION OF HYDROGEN AT A PRESSURE OF 1000 ATM

○ DATA CALCULATED IN THIS STUDY  
X DATA REPORTED IN REF. 2



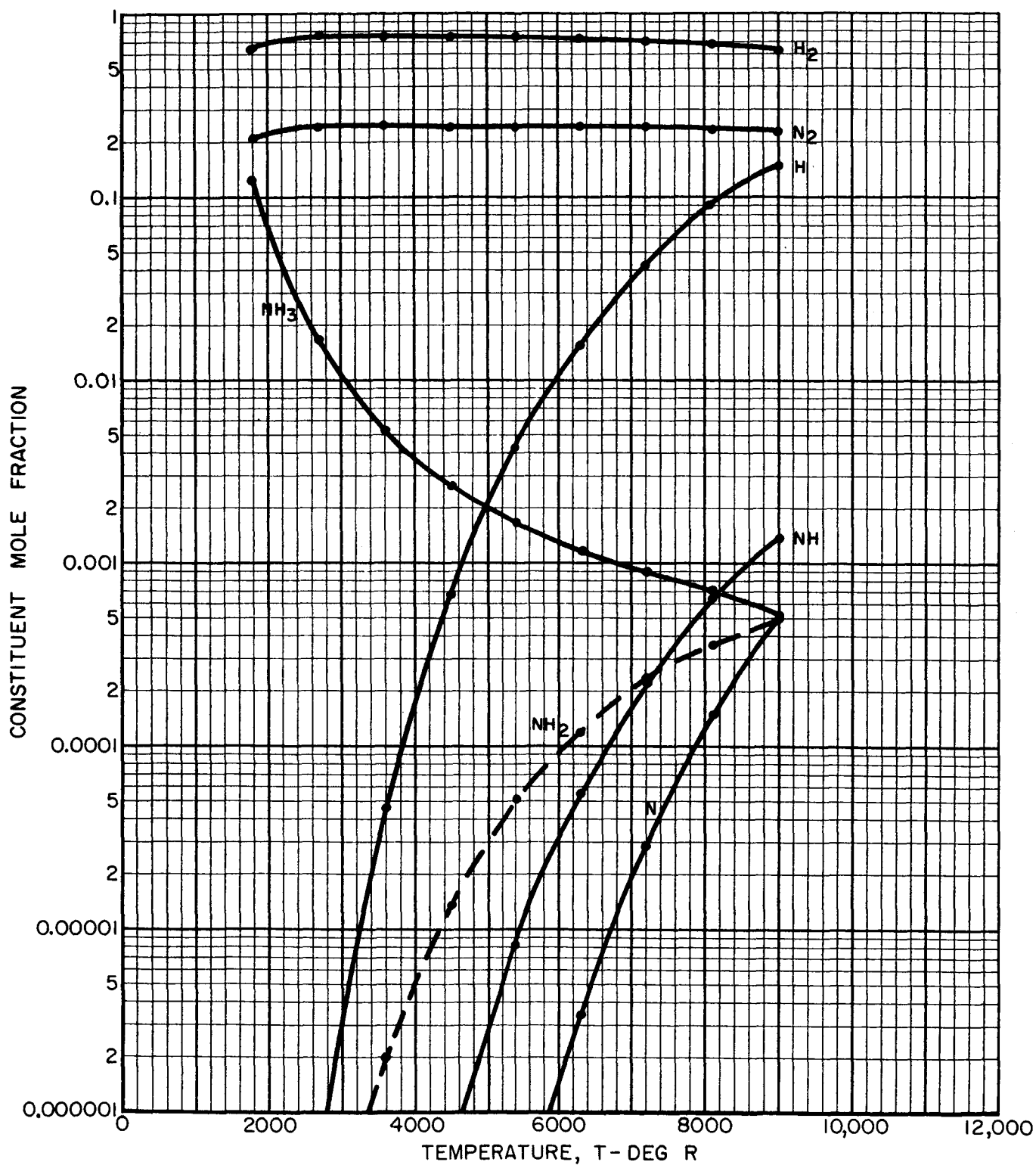
# EFFECT OF TEMPERATURE ON THE COMPOSITION OF METHANE AT A PRESSURE OF 1000 ATM

SYMBOLS INDICATE CALCULATED POINTS



# EFFECT OF TEMPERATURE ON THE COMPOSITION OF AMMONIA AT A PRESSURE OF 1000 ATM

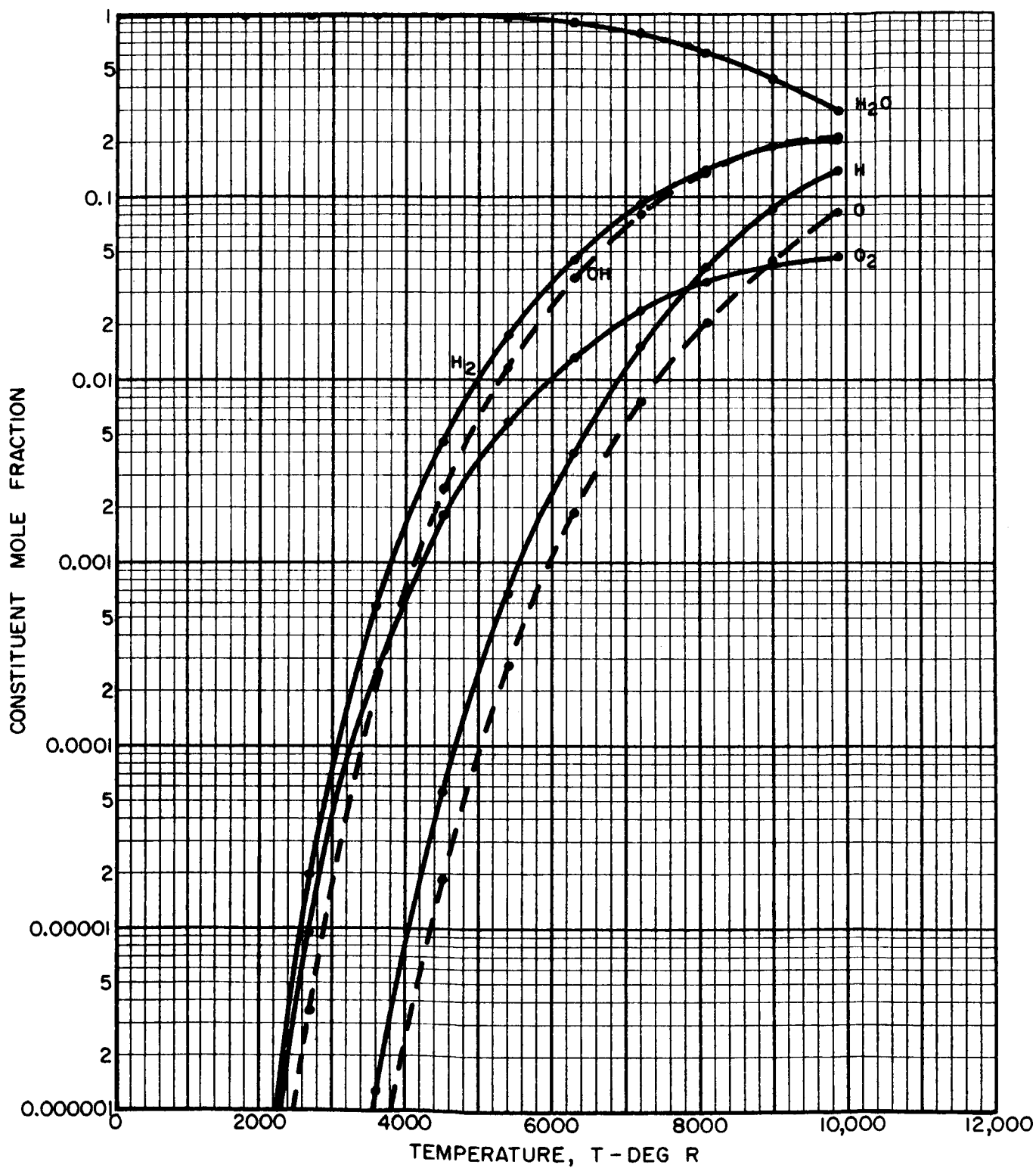
SYMBOLS INDICATE CALCULATED POINTS





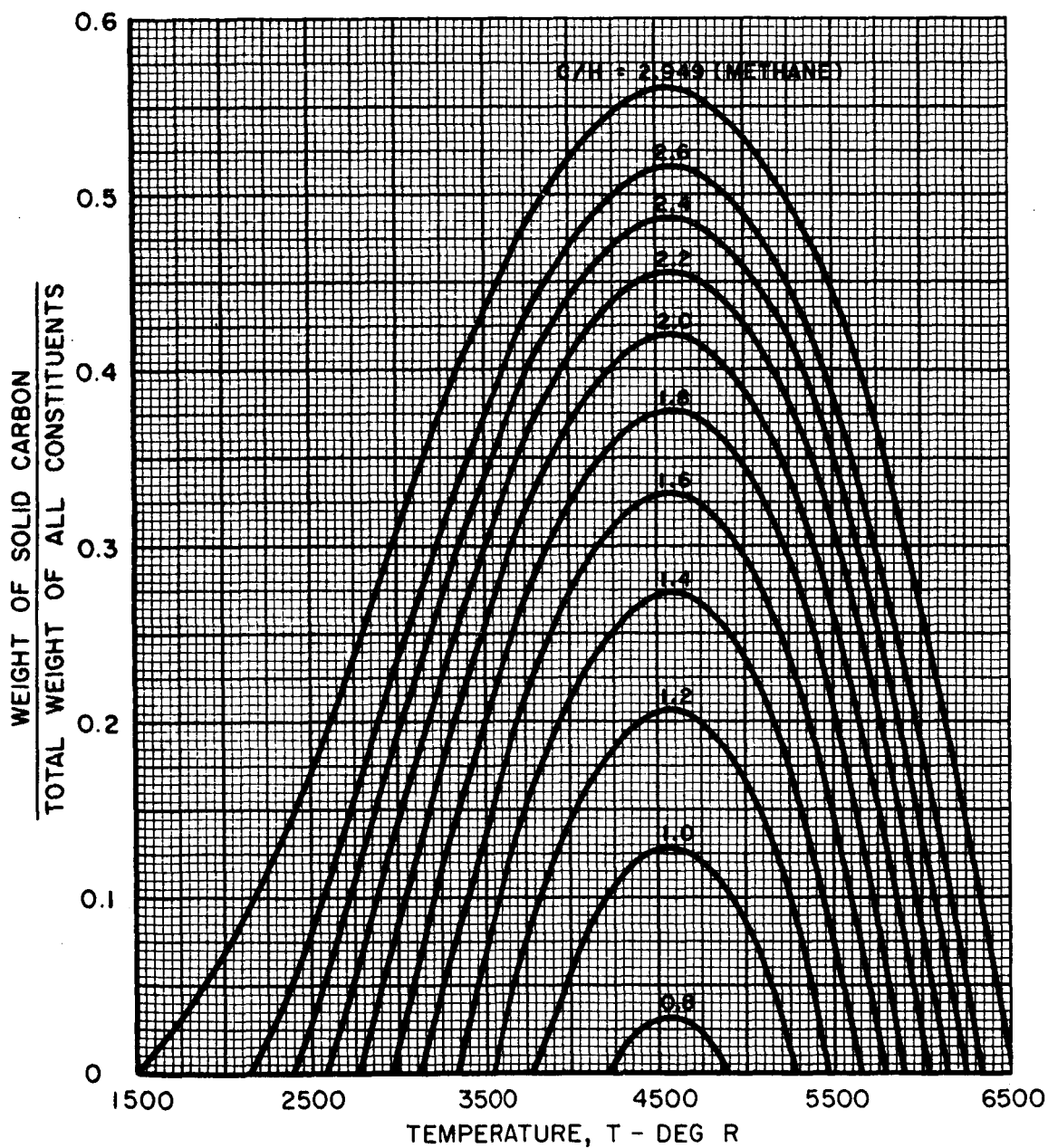
# EFFECT OF TEMPERATURE ON THE COMPOSITION OF WATER AT A PRESSURE OF 1000 ATM

SYMBOLS INDICATE CALCULATED POINTS



VARIATION OF SOLID CARBON CONCENTRATION  
WITH PROPELLANT CARBON-TO-HYDROGEN  
WEIGHT RATIO

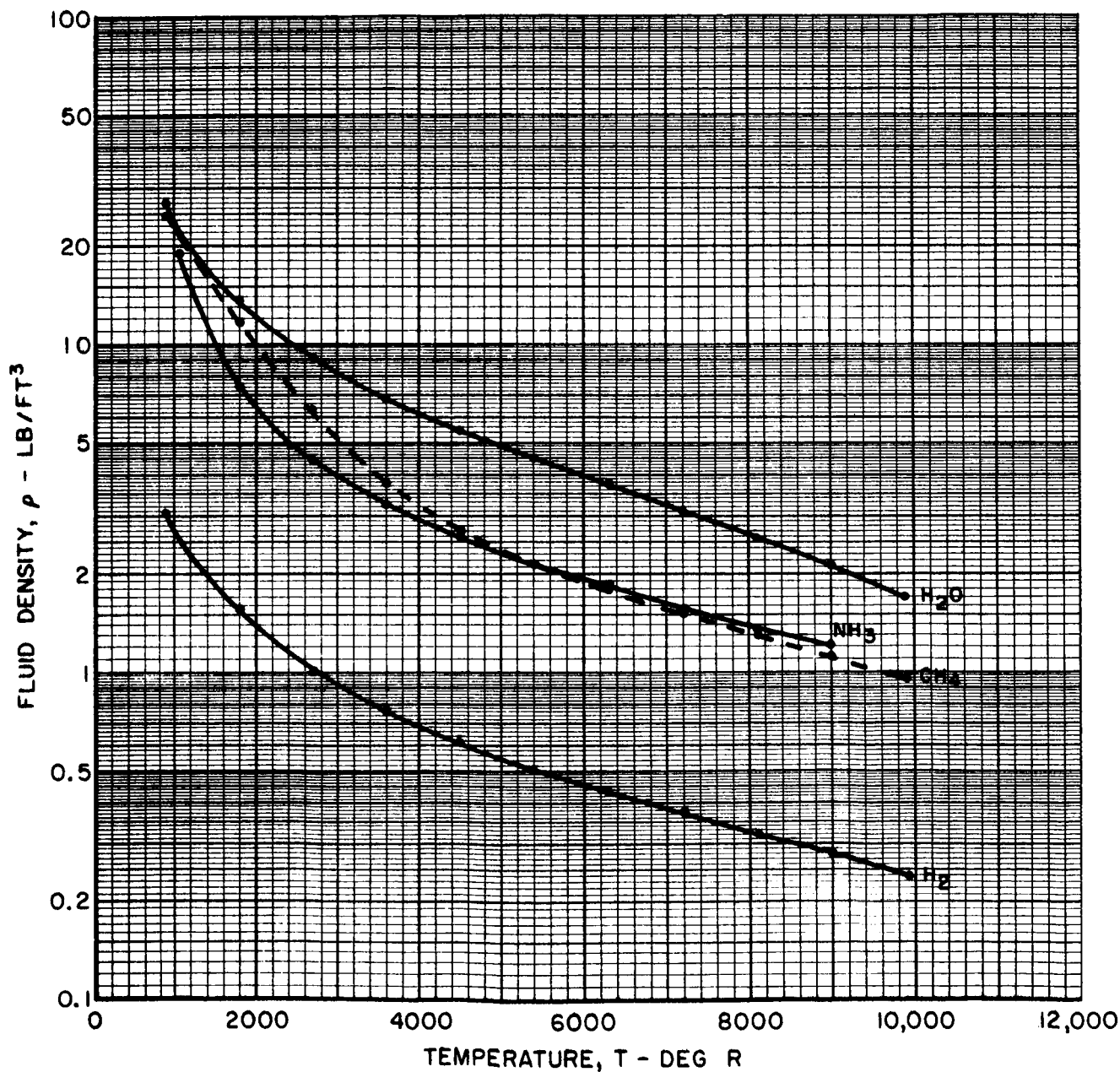
P = 1000 ATM



# EFFECT OF TEMPERATURE ON DENSITY OF WORKING FLUIDS AT A PRESSURE OF 1000 ATM

CALCULATIONS INCLUDE EFFECT OF DISSOCIATION

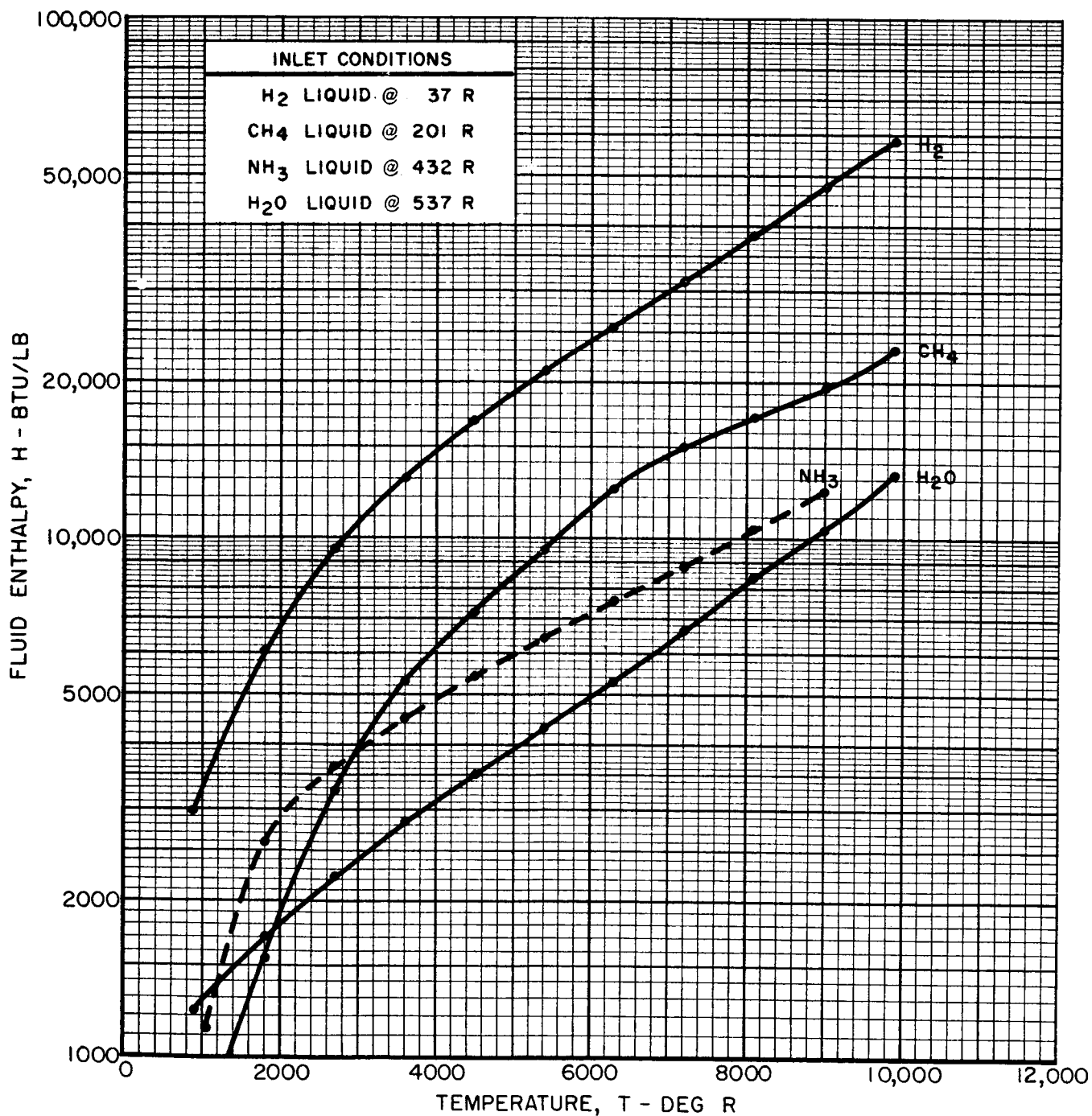
SYMBOLS INDICATE CALCULATED POINTS



# EFFECT OF TEMPERATURE ON ENTHALPY OF WORKING FLUIDS AT A PRESSURE OF 1000 ATM

CALCULATIONS INCLUDE EFFECT OF DISSOCIATION

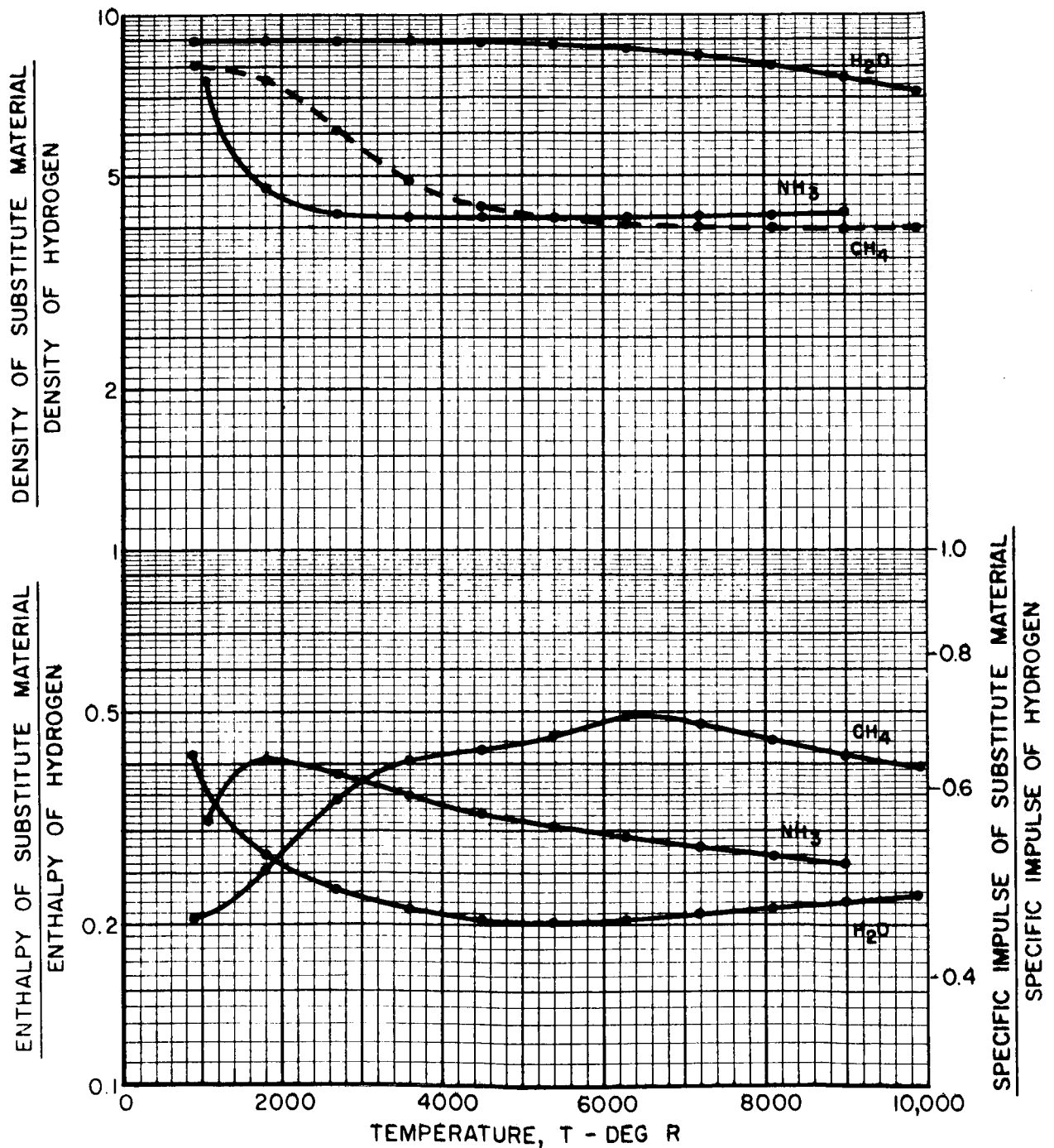
SYMBOLS INDICATE CALCULATED POINTS



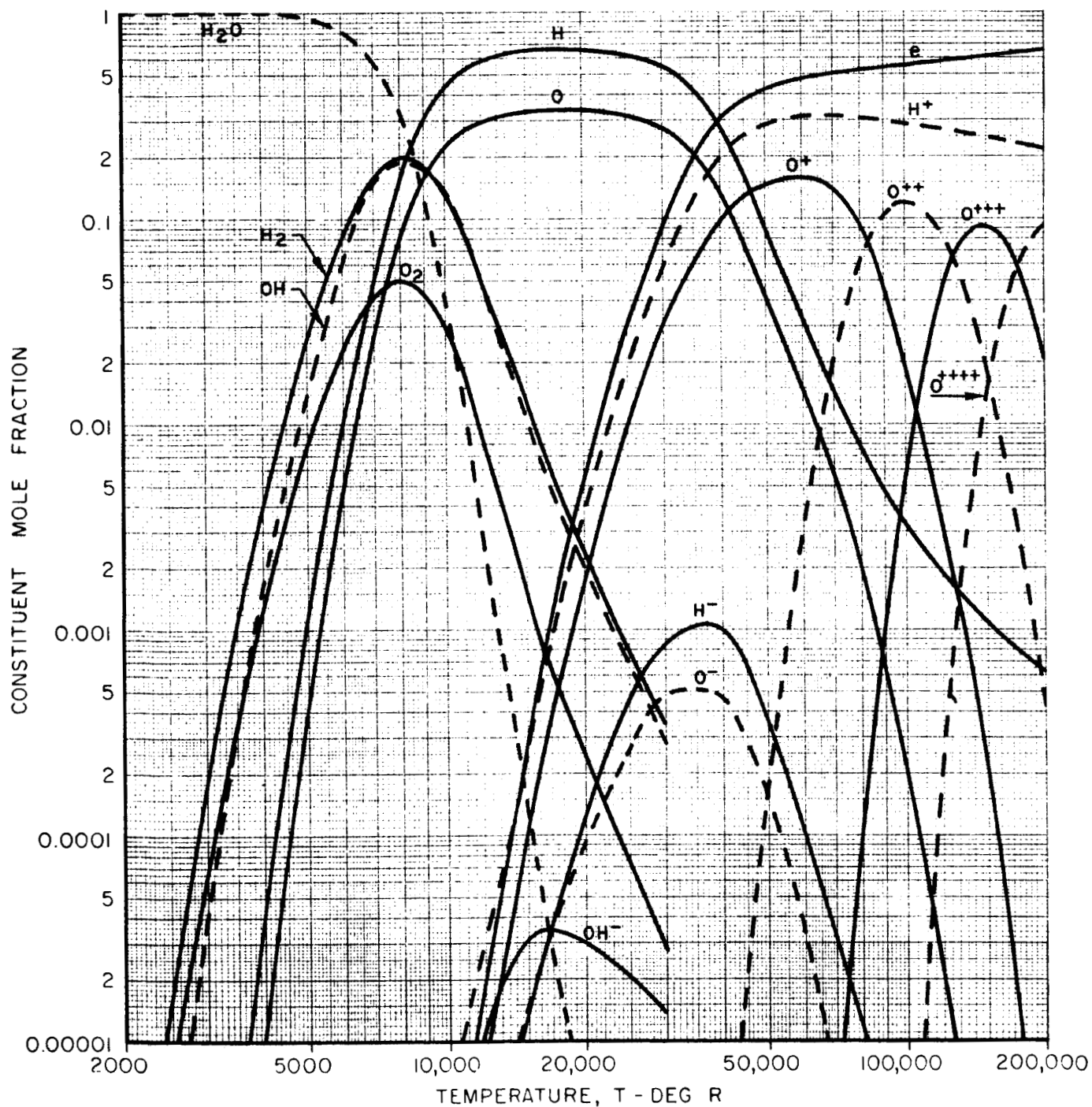
# EFFECT OF TEMPERATURE ON THE RATIOS OF SELECTED PROPERTIES OF SUBSTITUTE MATERIALS TO THOSE OF HYDROGEN

$P = 1000 \text{ ATM}$

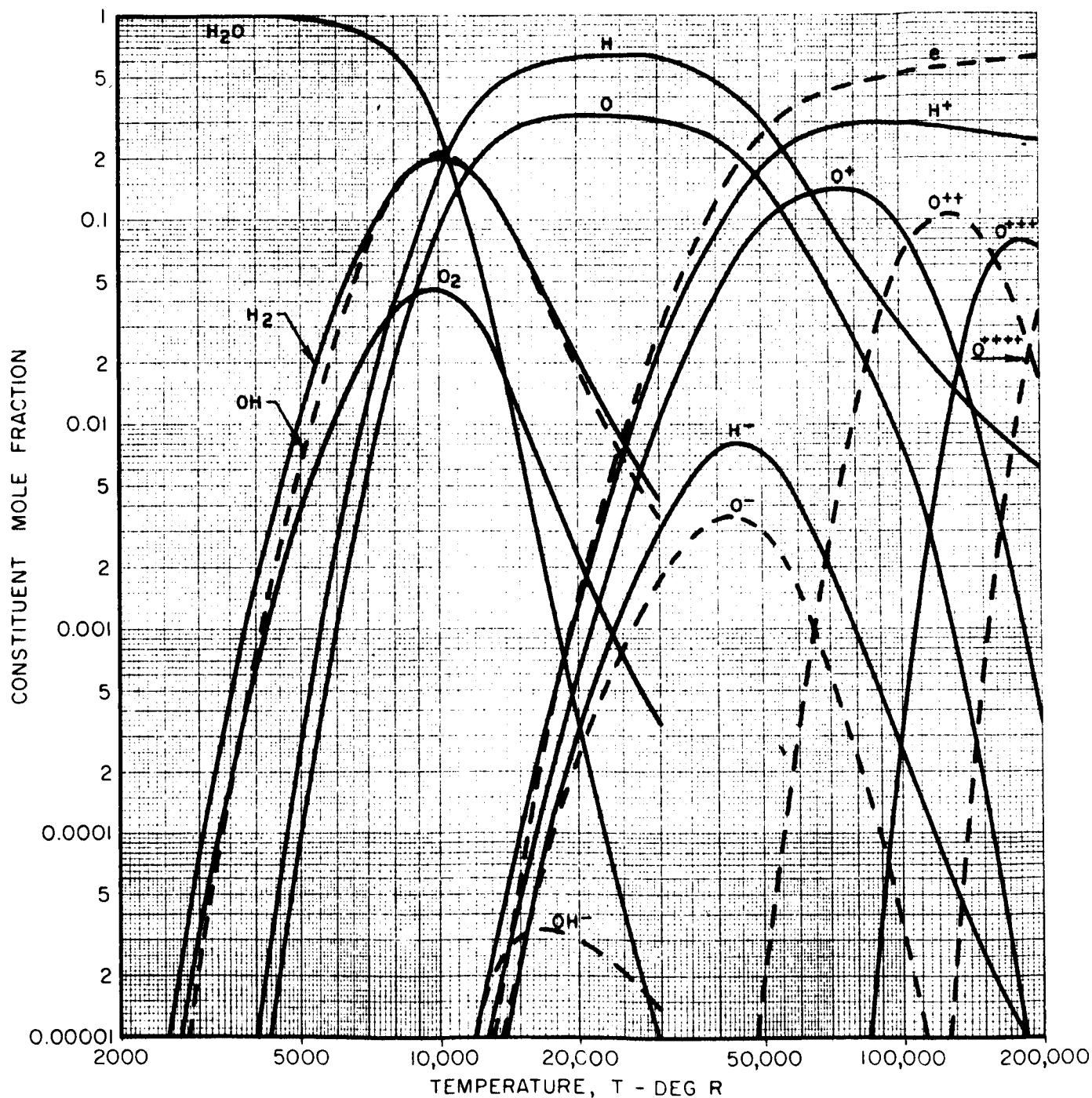
SYMBOLS INDICATE CALCULATED POINTS



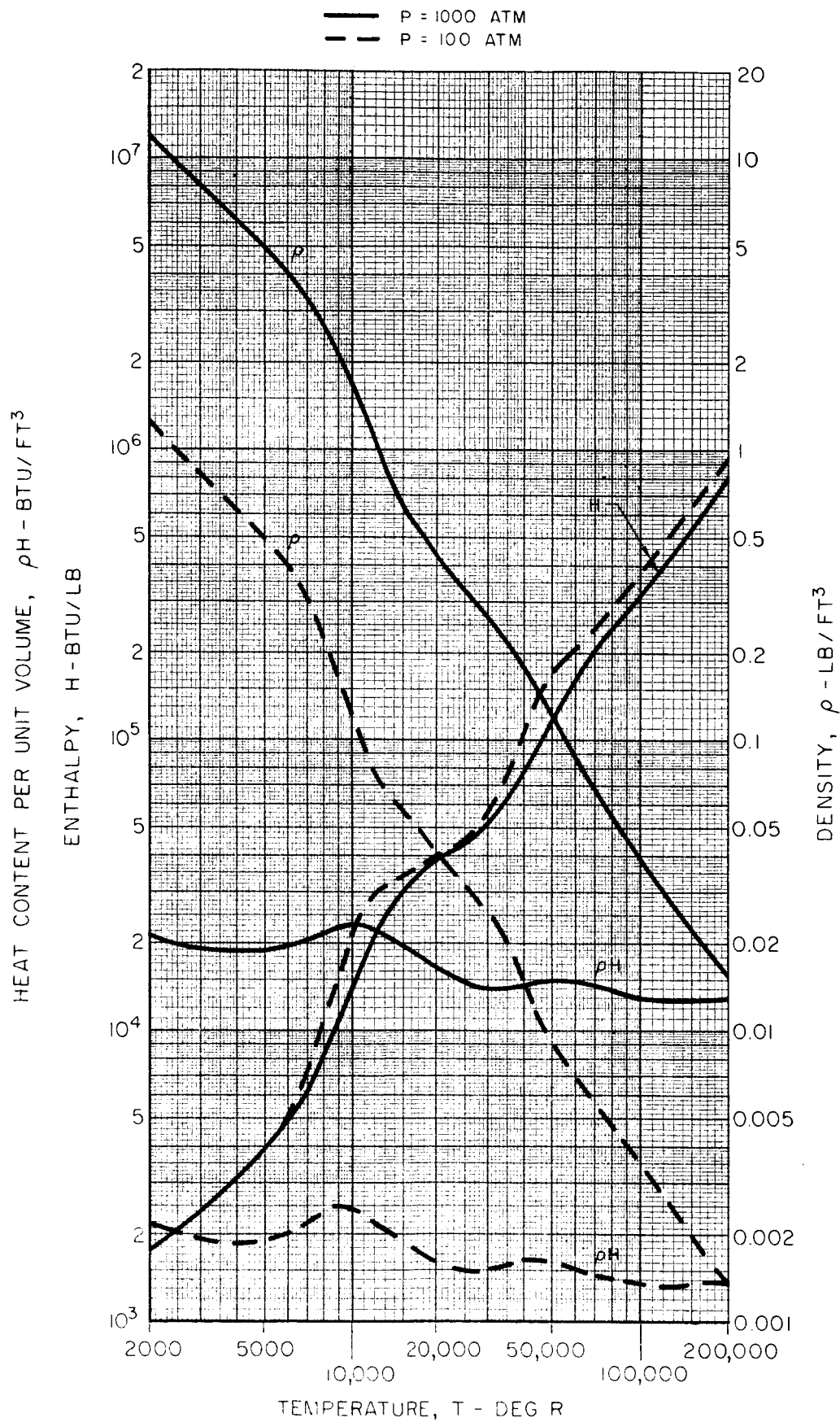
EFFECT OF TEMPERATURE ON THE  
COMPOSITION OF WATER AT A PRESSURE OF 100 ATM  
FOR TEMPERATURES TO 200,000 R



EFFECT OF TEMPERATURE ON THE  
COMPOSITION OF WATER AT A PRESSURE OF 1000 ATM  
FOR TEMPERATURES TO 200,000 R



# EFFECT OF TEMPERATURE ON ENTHALPY, DENSITY, AND HEAT CONTENT PER UNIT VOLUME OF WATER





# CONCENTRATION OF CONDENSED METAL SEED PARTICLES

P = 1000 ATM

CONCENTRATION OF SEED MATERIAL = 2% BY WEIGHT OF HYDROGEN-SEED MIXTURE

NUMBER IN PARENTHESES INDICATES MELTING TEMPERATURE IN DEG R

